

1999 AS A SPECIAL SPATIAL YEAR FOR PCBs IN HUDSON RIVER FISH

by

Ronald J. Sloan, Michael W. Kane and Lawrence C. Skinner

Bureau of Habitat, Division of Fish, Wildlife and Marine Resources
New York State Department of Environmental Conservation
Albany, New York

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Abstract: Long term monitoring of PCBs in fish from the Hudson River has occurred since 1977 and the temporal trends from specific locations (river reaches) have been widely reported. In response to a recognized need to more fully evaluate PCB concentrations arising from more localized source conditions, a greatly expanded sampling project was undertaken in 1999. The data were expressed on both wet weight and 'lipid' based concentrations, but most discussion focuses on the lipid adjusted values. Similar to earlier work, the spatial gradient away from the predominant PCB source area near Hudson Falls was evident. A strong association of localized PCB sources related to major PCB deposits and discharges was found as well as the evidence of other source conditions related to much smaller, but locally significant, inputs. The results emphasize the need to better evaluate and scrutinize the potential impacts of smaller, localized pockets of contamination. The source conditions included known sources such as the Queensbury site above Glens Falls, the original discharge points from the GE capacitor plant sites in Hudson Falls and Ft. Edward, the contributions from the Thompson Island Pool below Ft. Edward, and the general area above the Federal Dam at Troy (the 'Upper River'). PCB concentrations in fish in the Upper River showed considerable heterogeneity. This area presumably reflects the conditions related to the PCB-laden sediments in this 40 mile reach of the river. Below the Troy Dam (the 'Lower River'), the PCB contaminated waste site at Hastings-on-Hudson was also readily discernible in the data. Other areas in the river which should be evaluated further include the mouth of Catskill Creek, the Shad Island area, the area of the Remnant Deposit sites along the shoreline near Ft. Edward, and the area immediately above Bakers Falls Dam at Hudson Falls.

The large numbers of samples, locations and species analyzed, permitted the use of a novel approach to evaluating data. The term 'species smash' describes a mathematical expression of all the PCB results averaged, usually on an individual basis, for all the samples collected for all the species (usually more than four), at any given location. The combined results provide an assessment for that location. The 'species smash' which can cross class and order lines is viewed as a powerful tool in evaluating bioaccumulable contaminant conditions for impacted sites in aquatic environments. In the final analysis, the resulting values for the 'smash' that were derived from the increased sample numbers, and number of species, collected in 1999 from 65 locations, allowed the fuller expression of spatial gradients with distance from a predominant source. Yet this approach showed sensitivity for differentiating localized influences. The data from 1999 indicate that all sources are localized. For any one source the influence may be large, for example the influence with the sediments of the Upper Hudson River. But at locations where another source exists, e.g., Hastings-on-Hudson, that more localized influence is also discernible in the biota. It is also possible to distinguish between sediment deposits within a given reach of the river, e.g., the area near SA13 versus the east channel around Rogers Island, or the channel behind Griffin Island, all of which are located in the Thompson Island Pool.

Other aspects of PCB contamination that were examined included a comparison of liver and muscle concentrations in several species including freshwater fish, striped bass, Atlantic

tomcod and blue crab. On a wet weight basis, there was a tendency for liver tissue to have higher concentrations compared to the muscle (edible) portions, but there were exceptions. However, expression of results on a lipid basis showed that the edible portion was comparable to, if not higher than, the concentrations in the liver. Examination of another limited portion of the 1999 data set for seasonal changes in contamination was not conclusive and indicated the need for additional studies.

There is also an apparent need to obtain lower detection limits, since some sites with concentrations at or less than the current detection limits produced results that are most likely inflated. PCB concentrations in some areas of the Hudson River may well be consistently less than 0.5 ppm on a lipid basis or at or below 0.05 ppm on a wet weight basis.

INTRODUCTION

In the 25 years following the recognition of the severity of the PCB problem in the Hudson River documented in 1974 by Nadeau and Davis (1976), there has never been an extensive evaluation focused on describing and comparing PCB conditions in the biota in different locations in the river. Spagnoli and Skinner (1977) presented a statewide perspective on PCB contamination, including a summary of the Hudson River data that were available at that time. Their data coupled with other investigations on water and sediment led Horn et al. (1979) to describe the Hudson River as the most highly PCB contaminated river in the USA. With the advent of the Hudson River Advisory Committee arising from the 1976 PCB Settlement Agreement between the State of New York and the General Electric Company (GE), funding was made available to support the Long Term Hudson River PCB Analysis Project (the Project). The GE discharges of PCBs to the Hudson River resulted in high levels of contamination in the fish (Sofaer 1976). The subsequent Project focused on documenting the temporal trends in PCB concentrations in selected species of fish from selected locations along the length of the river from above Glens Falls to New York City.

The Project has described, through time, the course of contamination over a 200 mile stretch of a free-flowing freshwater stream which also featured, in the lower stretches below river mile 150, fresh to brackish tidal flows. The trends in the contamination are described in several publications (e.g., Sloan et al. 1983, 1984, 1988, 1995; Armstrong and Sloan 1988, Sloan and Armstrong 1988, Brown et al. 1985, Horn and Sloan 1985, Sloan and Horn 1986, Sloan and Hattala 1991, Sloan and Field 1996; Sloan 1993, 1994, 1999a, 1999c; USEPA 2002). In evaluating long term trends in PCB for several areas in the Hudson River, it became apparent that even highly mobile migratory fish species could exhibit contaminant concentration conditions related to localized sources. Sloan et al. (1995) discussed the ability to distinguish between locations based on the PCB concentrations even for an anadromous, migratory species like the striped bass.

In 1999, a special supplemental project to the Long Term Hudson River PCB Analysis Project was implemented by the New York State Department of Environmental Conservation (NYSDEC), Division of Fish, Wildlife and Marine Resources (DFWMR) with the support of the Division of Environmental Remediation (DER). The principal objective was "To evaluate the spatial relationships of contaminant concentrations in fish and to relate the results to source conditions, in so far as possible."

Additional objectives included examining the results to determine the advisability of more extensive sampling in subsequent years, or collecting from other potential source locations. As the collections progressed, interest developed in evaluating differences in organ concentrations, principally liver compared to the edible or standard fillet portions, and documenting seasonal

changes in concentrations at selected locations. Since other parties, including the General Electric Company and the National Oceanic Atmospheric Administration (NOAA), expressed interest in the results, there were opportunities for inter-laboratory comparisons of the PCB results on split samples. As is done with all data collected by DFWMR on contaminants in biota, the information was utilized by the New York State Department of Health (NYSDOH) in updating advisories on fish consumption which are made public through publication and dissemination of advisory information through various outlets such as the booklet, "Chemicals in Sportfish and Game: 2001-2002" (NYSDOH 2001).

METHODS AND PROCEDURES

Any organism collected, mentioned, or discussed in this paper and included in the 1999 sampling effort is listed in Table 1, which provides accepted common and scientific names down to genus and species, where possible.

In the Long Term Hudson River PCB Analysis Project (the Project), there are 10 general localities targeted for collection. But because there are three separate project elements (yearling pumpkinseed, adult resident species and striped bass), the annual collections actually originate from 15 specific locations. These sites include: Above the Feeder Dam near Glens Falls for adult fish and yearling pumpkinseed, Thompson Island Pool (behind Griffin Island for adult fish, and east side of river near Griffin Island for yearling pumpkinseed), Stillwater Pool at Coveville (adult fish), above Stillwater Dam for yearling pumpkinseed, below the Federal Dam at Troy for adult fish including striped bass, south turning basin at the Port of Albany for yearling pumpkinseed, Catskill area for adult fish and striped bass, Poughkeepsie area for adult fish and striped bass, above Marist College in Poughkeepsie for yearling pumpkinseed, Newburgh area for adult fish, south end of Denning Point for yearling pumpkinseed, Stony Point area for striped bass, Piermont area for adult fish and striped bass, and near the George Washington Bridge for striped bass.

The original location list for the 1999 supplemental collections focused on 27 sites, but the list was augmented as the work progressed. These sites were in addition to the locations normally sampled in the Project. The final list of all the collection sites in 1999 is provided in Table 2. The locations are plotted in Figure 1. The Federal Dam at Troy (RM 153) is a geographic feature which separates the Hudson River into two 'Upper' and 'Lower' sections. The Upper Hudson River, which includes the confluence with the Mohawk River at Cohoes at about RM 154, is largely a free flowing riverine system. However, the 40-mile reach from Troy to Ft. Edward (about Rm 193) is further characterized by a series of seven navigational dams and locks which produces relatively quiescent conditions behind the impoundments. The Lower River below the Federal Dam to New York City is a 150 mile tidal estuary with a salinity gradient ranging from fresh to nearly saline.

The targeted number of samples was 45 fish from each location for the supplemental collections. At each location up to nine species represented by five fish per species across an array of legal/edible sizes were sought. The final numbers and species collected, along with PCB results, are provided in several summary tables.

Some locations involved assessing older PCB deposits associated with the operations of the GE Hudson Falls and Ft. Edward Plant sites. In these areas, known as the Remnant Deposits, materials were left behind after the removal in 1973 of a deteriorating log crib dam in the Village of Ft. Edward. Since these locations are severely restricted in size, collections focused on invertebrates, along with juvenile fish and minnow species. Some of the other collection

locations, such as those associated with the Hastings-on-Hudson waste site, also necessitated collecting invertebrates and smaller species/sizes of fish, due to habitat limitations.

In cooperation with NOAA, some samples were collected to evaluate seasonal changes in PCB levels on a congeneric basis, and to compare PCB concentrations in standard fillets versus levels in the liver. The seasonal evaluation focused on limited collections of largemouth bass, white perch and yellow perch from three locations - Newburgh (RM 60), Catskill (RM 113) and Coveville (RM 176). Samples for the liver analyses were taken from Catskill (RM 113), below the Federal Dam at Troy (RM153), Coveville (RM 176) and Griffin Island (RM 189) for selected species - brown bullhead, largemouth bass, striped bass, yellow perch and white perch. Not all the species were collected at each location but the target sample size was five fish for any particular species/location combination. Spring samples were collected in late May and early June and the fall samples were taken in late September and early October. Liver samples were taken from spring collected fish. A special collection of Atlantic tomcod occurred in January and February 2000 which also involved liver tissue analyses.

Methods of collection varied but most efforts utilized an 18 foot electrofishing boat (Smith-Root model SR18E) equipped with a variable output 900 volt gas-powered DC generator. The aluminum vessel was powered with a Mercury 140 horsepower jet engine and had a sufficiently shallow draft to allow working in water depths at 0.5 meter or less, when necessary. Operating amperage was maintained between 7 and 12 amps depending upon the conductivity of the water. Invertebrates were taken by handpicking, small seines, dip nets, shovels and buckets. Small fish and areas inaccessible to the shocking boat were sampled with seines, gill nets, backpack electroshocking and angling techniques.

Fish and invertebrate samples were handled according to standard DFWMR procedures (Sloan 2000) which entail recording on standard forms for each specimen, the date of collection, a unique identification number or code, the location including GIS coordinates, species (genus and species, if possible), length in millimeters, weight in grams, sex (if possible), and method of collection. Chain of Custody forms were maintained and samples kept cool and then frozen on the same day of collection.

Samples were later processed by experienced personnel at Northeast Analytical Laboratory (NEA) in Schenectady, New York or the New York State Department of Environmental Conservation (DEC) laboratory, Hale Creek Field Station (HCFS), in Gloversville, New York. Frozen prepared tissues, as either standard fillets, specific organs/tissues, or whole bodies, were then shipped overnight via air freight to the DFWMR contract laboratory, Mississippi State Chemical Laboratory (MSCL), Mississippi State, Mississippi, for PCB and lipid content analyses. Some portions of the collections were analyzed at the HCFS laboratory. Selected fish were also analyzed as split samples for PCB congeners at NEA, funded by the General Electric Company, or at Severn Trent Laboratories (STL) in Colchester, Vermont, at the request of, and funded by, NOAA.

Analyses for PCBs as 'Aroclors' involved at least a seven hour Soxhlet extraction with hexane to remove the lipid material, which was then prepared for gas chromatograph (GC) determinations, according to MSCL or HCFS standard operating procedures (SOPs), to quantify the estimates of the PCBs in the samples. The extractable portion of the original mass of fish tissue, expressed as a percent, is used to represent the fat content of the organism and is referred to as 'lipid content.' Non-detect values were usually treated as ½ the detection limit for a given 'Aroclor.' Detection limits were, for each 'Aroclor', 0.01 ppm at MSCL and 0.02 ppm at HCFS. Providing data as 'Aroclors' allowed the partitioning of the results into lower chlorinated components and a higher chlorinated fraction, thereby permitting rough approximations of PCB composition into two classes, higher versus lower chlorinated PCB forms. Both laboratories modified the 'Aroclor' quantitations to minimize the potential influence of double counting overlapping peaks between closely related mixtures, either through modified calibration steps or the algorithms used in estimating concentrations.

Congeneric analyses were conducted similarly, but the procedures relied on individual peak identification and quantitation from separations on capillary columns with an electron capture detector (ECD) equipped GC.

Data were collected and stored on Dell PCs per the data dictionary (metadata) developed by the Bureau of Habitat, DFWMR, Albany, New York in a Visual FoxPro® version 6.0 database format. Linked files were queried for summarizing the data and conducting statistical analyses using Excel® and Statgraphics Plus® software.

RESULTS AND DISCUSSION

The Collections

From April 16 through November 19, 1999, a total of 8641 organisms (5110 invertebrate and 3531 fish) were collected, which resulted in 2544 analyses for PCBs. Invertebrate PCB analyses totaled 218 and fish 2326. Because the opportunity arose early in 2000, sampling for fish at Sanford Lake in the headwaters of the mainstem Hudson also took place and provided data on PCBs in biota associated with an area which is not considered directly contaminated by PCBs. Those results and all others generated for 1999 are included in the totals above and are compiled in Table 3 in summary form primarily for fish. Some of the invertebrates collected at locations other than the Remnant Deposits are also included. The table lists average PCB concentrations on both a wet weight and a lipid basis for each species collected at a particular location along with summary body measurement information for the collection. Table 4 provides summary information for the invertebrates collected as part of the evaluation for the Remnant Deposit area near Hudson Falls and Ft. Edward. Figure 1, appearing as six (6) different views, shows the collection locations from Lake Sanford, river mile (RM) 301 in the headwaters of the Hudson River to the George Washington Bridge in New York City at RM 12. Table 2 lists the specific locations with a brief description along with the river mile assigned to that position for the purposes of data analysis and interpretation. The river miles as given are intended only as approximations to allow differentiating between points. The names of the organisms collected in this project are listed in Table 1 by acronym (species code) as they appear in the Bureau of Habitat master database along with the accepted common name and/or a description if a common name was not known, and the scientific name or a technical designation such as a family or genus.

A special collection of Atlantic tomcod was arranged through the New York Power Authority for the winter of 2000. Those results are summarized in Table 5.

Laboratory Comparisons - "Aroclors" versus Congeneric Analyses

For several years, the Northeast Analytical Laboratory (NEA) of Schenectady, New York, funded by GE, has conducted analyses on some of the same samples analyzed by the DEC contract laboratory. The analytical approach used by the laboratories differed in that NEA quantified the results on a congeneric basis, whereas the DEC contract laboratory, Mississippi State Chemical Laboratory (MSCL), estimated the quantities of PCB on an 'Aroclor' basis. The latter method is the historical option for analysis, and is still in use since the US Food and Drug Administration tolerance level is based on the 'Aroclor' procedure. The vast majority of the data developed over the last 30 years is in the 'Aroclor' form.

Table 6 and Figures 2 and 3 provide comparisons between the two analytical methods. Two species, largemouth bass and brown bullhead, from two locations, Thompson Island Pool

and Stillwater, were analyzed in 1999 by the NEA and MSCL laboratories using ground homogenized subsamples of the same fish. The results showed no significant differences on average for total PCB, either on a wet weight basis or lipid basis, even though the methodologies were quite different. A third species, yellow perch, showed similar results but it is not graphed separately, although the data are summarized in Table 6.

Severn Trent Laboratory (STL) also produced results on 103 split samples on a congeneric basis. Since these analyses represent smaller sample sizes from more locations than the NEA comparisons, the relationship between MSCL and STL appear in Figure 4 as scatterplots and regressions for all the results available. A summary for all of the STL data are provided in Table 7. The number of fish in the table is different from the number of split samples noted above because the mass of tissue used in the original analysis conducted by STL left no sample available to MSCL for analysis. The graphical comparisons are based only on actual 'split' samples. In the development of the regressions for Figure 4, if a different association, i.e., logarithmic or exponential compared to a linear expression, improved the fit, it was chosen over the linear equation for presentation in the graphs. The relationship for total PCB between the two labs is better on a wet weight basis (Figure 4-A) than it is on a lipid basis (Figure 4-B). Figure 4-C shows the association between the laboratories in estimating lipid content, and reflects a relatively greater degree of variability than is apparent in the wet weight PCB association (Figure 4-A). Hence, the R^2 is lower for the lipid based PCB association and the expressed concentrations on a lipid basis tend to be higher since STL appears to have removed less lipid material than did MSCL. For either lipid based or wet weight values, as concentrations increase, there is tendency for greater variability even though the correlations between the laboratories remain high. In any case, there is obviously no direct 1:1 relationship between the laboratory results since none of the linear models produced higher R^2 's than the exponential fits.

Since STL had the first opportunity to analyze the samples, some of the potential for introducing artifacts into the process as it may relate to changes in storage, preparation and shipping conditions were minimized. Although it is not certain, efficiency of extraction for PCB or lipid material may also be more variable, or less consistent, at higher levels as seen to some degree in Figure 4-C and STL may have used a separate step in determining lipid content that utilized a different solvent, methylene chloride, rather than a 50:50 mixture of acetone/hexane which was the solvent mix for the Soxhlet extraction procedure. This may have contributed to relatively lower lipid contents for STL. Figure 5 presents a comparison of the overall averages on both wet weight and lipid bases between the two laboratories, STL and MSCL. Even though the average differences are not large, there is a shift in the difference between lipid based and wet weight concentrations. MSCL is slightly higher on a wet weight basis but STL is higher on a lipid basis again indicating that STL may have removed lesser amounts of lipid.

However, all three laboratories, MSCL, STL and NEA, generally produced comparable results for total PCB. The congeneric analytical results from STL and NEA, and the associated complexities they present, are beyond the scope of this paper. It is important to this interpretation, however, since these more complex analyses indicate that there can be general

agreement between different methodologies in the determination of total PCB.

Influence of Age/Size/Sex/Lipid Content on PCB Concentration

Several factors are commonly felt to influence contaminant concentrations in biota. For several species from some locations for which the samples sizes were relatively high, the correlations between length, weight, age(where available), and lipid content to total PCB are presented in Table 8. Of the 32 species-location combinations for which the sample sizes available would allow the derivation of a correlation matrix with some confidence, 17 cases were significant ($P < 0.05$ or $P < 0.01$) for the relationship between total PCB and percent lipid. The correlations were all positive. Eleven (11) of the remaining species/location combinations were positive or had higher coefficients, although not significant, for PCB and lipid compared to the other species/location combinations. There were eight (8) cases where the correlation of PCB with length was significant but two of these were significantly negative. Similarly, in nine (9) cases, PCBs were correlated with weight, but two of these were significantly negative. Six of the positive significant correlations of PCB with length and/or weight involved the same cases in which there were significant correlations with lipid. Available age data were insufficient to prove useful as a variable in evaluating the relationship between age and PCB content (Table 8). However, given the tendency for lipid content to exhibit a better relationship with PCB concentration than length or weight, age may not be a useful descriptor for accumulation either.

Age/Size versus PCB - Nevertheless, some fish were aged such as the Atlantic tomcod from the Lower River collected in January and February 2000, the pumpkinseed collected as part of the yearling pumpkinseed project, and samples of several species from the Sherman Island Pool involving the Niagara Mohawk Queensbury PCB Project. The Atlantic tomcod (Figure 6 and Table 5) do not exhibit age differences in the level of PCB contamination but this species is discussed further in the section on liver-standard fillet relationships (page 21). The excess pumpkinseed taken at and above Marist College at Poughkeepsie provided a range of ages to evaluate the relationship between age and PCB content. There was a significant correlation in the collection at Marist College but not at the site upstream from the college, although the trend in the data was positive (Table 8).

The results from the Niagara Mohawk Queensbury site (NiMo1) are of the greatest interest in interpreting age/size contaminant relationships. The site is a small hazardous waste site located in the impoundment formed by the Sherman Island hydroelectric power dam. It is a focus for remediation of PCBs in an underwater exposed sediment portion of about eight acres in size. In order to evaluate impacts to biota, five sampling locations were established to characterize and evaluate the extent of contamination as shown by fish (pages 10 and 11 of Table 3, Figure 1-C). Long term monitoring of the fish shows that the removal of much of the PCB source conditions by 1996 resulted in dramatic reductions in the concentrations observed in the biota within the next year (Parsons 2001) which enabled the removal of fish consumption advisories for the Sherman Island Pool (NYSDOH 1998).

To evaluate the influence of age on subsequent accumulation of PCBs at the site, refer to the yellow perch data detailed on pages 10 and 11 of Table 3. The smallest specimens collected at the Queensbury site after the sediment remediation were analyzed for PCBs, but were not aged. These fish had the higher concentrations, on average, (at 9.81 ppm wet weight and 501.7 ppm lipid basis) compared to the larger fish, which were aged. The larger four and six year old fish contained 0.13 and <0.05 ppm, respectively on a wet weight basis. On a lipid basis, the two ages had 17.7 and 5.5 ppm, respectively. Since the smaller fish have less total energy requirements and the available resources in the vicinity of the site can meet their ecological needs, there is no advantage to forage over wider areas and staying localized has survival advantages in that predation is lessened. The larger fish, on the other hand, are forced to forage more widely. As a result, their exposure to the source condition is reduced. By obtaining their energy from other less contaminated locations the concentrations they exhibit are decreased. The contamination they received earlier in life is diluted through growth derived from prey taken in uncontaminated areas. This general phenomenon has also been noted for striped bass in the Hudson River but on a larger geographic scale (Sloan et al. 1995).

Although the other aged species (rock bass and smallmouth bass), also show a similar reduction of concentration with age, the sample sizes to fully describe the relationship were not available. At other locations in the Sherman Island Pool, particularly across the river but adjacent to the contamination (NiMo 2, on page 11 of Table 3), the concentrations for the most part are near the detection limits and so the age relationship is somewhat moot. This age gradient is one manifestation of what may be considered as inherent patchiness in the system which is the focus of much of the remaining discussion in this paper. This patchiness is interpretable using the information provided by the fish and invertebrate samples and is explored in a geographical context in the section on 'Source Conditions.'

Sex differences versus PCB - Eleven species-location combinations produced enough samples to reasonably attempt a comparison of PCB concentrations by sex. Only two of these were significant at $P < 0.05$: yellow perch females from Coveville (Stillwater Pool) at 274 ppm on a lipid basis compared to males at 155 ppm; and spring collected striped bass in the lower Hudson River below Poughkeepsie had males with 34.3 ppm versus 21.2 ppm on a lipid basis for females. Fall collected striped bass from the Lower River did not show sex differences ($P > 0.05$). The species and locations which did not exhibit sex differences were brown bullhead above the Feeder Dam; brown bullhead at the pumphouse above Bakers Falls; brown bullhead, yellow perch and largemouth bass from Griffin Island in the Thompson Island Pool; yellow perch from Stillwater; smallmouth bass from below the Federal Dam at Troy; and white perch from above Marist College at Poughkeepsie. In general, these results show that it is not necessary to differentiate between the sexes in describing the spatial conditions based on the PCB concentrations in the biota.

Lipid relationship - In general, the association between PCB concentrations and lipid content has received much attention in the Hudson River (Armstrong and Sloan 1988, Sloan and Armstrong 1988, Sloan et al. 1983, Sloan et al. 1984, Brown et al. 1985, Sloan et al. 1995, Jones and Sloan 1989). In other systems and situations it is not always significant but does provide a generally positive association (Stow et al. 1997) and is useful in describing spatial patterns of PCB in fish. Since lipid determinations are simply the percent of hexane soluble materials generated during extraction of tissue samples, the characterization of the residue as 'lipid' is, perhaps, a loose interpretation of fat content. Another term, 'total organic extractables,' has been suggested to replace the use of 'lipid.' However, for simplicity in this paper, the word 'lipid' is retained. Since composition of lipid constituents and lipid content varies widely between and within species (Henderson and Tocher 1987), the association between PCB and lipid is not always isometric. Hebert and Keenleyside (1995) explored alternative means to better explain the variability of the contaminant-lipid relationship through the use of covariance to control for other variables that may also influence the association. Stow et al. (1997) indicated the influence of spawning condition on the relationship between PCB and lipid. Generally, during the spawning period in five species of salmonids there was good positive agreement but during non-spawning periods there was not good agreement. That there are discrepancies in whether lipid content can explain all or most of the variability in PCB content is not surprising since Henderson and Tocher (1987) describe many of the changes in lipid content and composition as a function of season, sex, age, spawning and other physiological states.

Since the association between PCB and lipid is not always a straightforward method to interpret PCB data for fish, and low correlations are often found, it is apparent that not everything is known about bioaccumulation. Since what is usually described as 'lipid content' is the residue removed from the tissue in the course of extraction, any derived relationship is perhaps simply correlative. Henderson and Tocher (1987), however, indicate that lipids evaluated for their biochemical and physiological attributes are usually extracted from tissues through the use of organic solvents such as hexane.

Regardless, the lipid adjusted data provide a reasonable approach to evaluating spatial relationships and allow interpretation between locations, which otherwise might not be possible. There are fish in the Hudson River which exhibit differences in fat content from one location to another, thereby necessitating a lipid based approach to evaluate concentrations at those sites (Armstrong and Sloan 1988). Expression of PCB concentrations on a lipid basis is more than a simple convenience to equalize the differences between species, taxa, time and space. Lipid based values are an important tool for data interpretation.

Although wet weight concentrations are undeniably important, most of the rest of this report will focus on lipid based values. Our conclusions will rely primarily upon the interpretation of lipid adjusted concentrations. In this context, the term lipid-normalization is not used since expression of PCB concentrations on a lipid basis may not improve normality at all. We have not examined the lipid based data in detail regarding normality, but have used the observation that

lipid content is correlated to some extent with PCB and our interest here is to describe in general terms the observed spatial patterns. Therefore, the more appropriate terms 'lipid based' or 'lipid adjusted' PCB concentrations are used. A comprehensive statistical evaluation of lipid PCB relationships is not the focus of this paper but as a topic it is admittedly of interest and needs further pursuit.

Concentrations over the Spatial Gradient

For any given species, contaminant concentrations generally decline with distance downstream or away from a source condition (e.g., Armstrong and Sloan 1988, Sloan and Armstrong 1988, Sloan et al. 1983, 1984, Sloan and Field 1996, Sloan and Jock 1990). The existence of the spatial gradient has been a principal feature of the PCB contamination in fish of the Hudson River (Sloan 1999a, 1999c; USEPA 2002, Field et al. 1996, Sloan and Field 1996) and provides the framework for the following discussion.

Single Species Examples - The usual procedure to examine the gradient is to focus on individual species. For example, smallmouth bass and carp collected in 1999 (Figures 7 and 8) show the influence of both the overall spatial gradient and local source conditions along the river.

In Figure 7 for the smallmouth bass, the fish at NiMo 1 still reflect the influence of the contamination at the Queensbury site even though the concentrations are much less than at other source areas. Further downstream there is a highly elevated average concentration at the GE pumphouse, which corresponds with the original historical discharge point for PCBs to the Hudson River from the GE Hudson Falls Plant. At the time of sampling, this particular location had undergone significant remediation in 1997 and 1998. It may be hypothesized that the fish were still exhibiting residual impacts and follow up sampling to track the efficacy of the remediation is in order. The concentrations in the bass through the east shore remnant sites and in the vicinity of the GE Ft. Edward plant 004 outfall appear greatly influenced by these sites, particularly around remnant 3 compared to remnants 2 and 4 on the west shore. Downstream of the remnant sites, concentrations increase substantially, starting in the upstream portions of Thompson Island Pool in the East Channel of Rogers Island and near the dredge spoil area known as Special Area 13 (SA13). From there downstream to Troy, PCB concentrations in the bass remain elevated with a particularly high concentration noted at the sediment depositional area known as *Hot Spot* 28. From Troy (RM 153) down to Constitution Island at river mile 54, concentrations in the bass are reduced but still substantial and noticeable.

Carp (Figure 8) were not available at the same locations as the smallmouth bass but they also exhibit the influence of PCB source conditions related to the sediments of the Thompson Island Pool and the Upper Hudson River down to Troy. From Troy downstream to Piermont Marsh, the PCB levels are lower, but still readily discernible. In the Upper Hudson River there is

considerable variation in PCB concentrations between locations; most notable is the Stillwater site where the average was dramatically influenced by one individual fish with over 2300 ppm PCB on a lipid basis. This illustrates the vagaries of having to rely on small sample sizes and single species in attempting to describe and understand contamination in a natural system, particularly in a stream as large and complex as the Hudson River.

Differences between Species - That differences in PCB concentrations between species exist in the Hudson River has been explored in various papers (e.g., USEPA 2000a, 2000b, Sloan et al. 1984, Sloan 1993, Sloan and Field 1996). Likewise, at a given location, such differences are readily apparent in Table 3 and it is of interest to explore this further in relation to trophic level. Commonly, brown bullhead are relegated by the public into the category of 'bottom feeder' and hence they are felt to be exposed to contaminants more so than other species. This gives rise to a particularly persistent, troublesome 'rufous harengus.' Brown bullhead, as do other members of the catfish family, actually exhibit more of an omnivorous, even opportunistic, feeding habit (Werner 1980, Smith 1985). Carp also are omnivorous and although they tend to feed on the bottom, they will often move up into the water column to feed. They do utilize more plant material than other species. Yellow perch are also generalists in their feeding but are oriented more toward insects, and will take fish of the proper size when available. They forage through different aquatic habitats including benthic and mid-column strata. Smith (1985) and Werner (1980) indicate the strongly carnivorous habit of the largemouth bass which take not only fish but almost any animal of suitable size that presents itself.

Generally, it is felt that the species tend to be more alike in average concentration at a given location than they are dissimilar. Figures 9-A, more contaminated locations, and 9-B, less contaminated areas, depict the fact that for their average PCB concentration, a particular species will shift position relative to the other species at a given location. There is generally a lack of consistency between locations in terms of position whether concentrations are expressed on a wet weight basis or a lipid basis. Carp may tend to be high in concentration on a wet weight basis and low on a lipid basis but it is not necessarily always the case. For example, brown bullhead in the less contaminated areas (i.e., in the downstream, tidal reach of the river), Figure 9-B, shift position relative to other species for average PCB concentration between Troy and Constitution Island. At Troy their PCB concentrations are comparable to largemouth bass and yellow perch (actually, intermediate to the two) but much lower than the carp on a wet weight basis. On a lipid basis they have the lowest average concentration. At Constitution Island, however, the brown bullhead produce the highest average concentrations compared to the other species on both wet weight and lipid adjusted bases. The species tend to shift their relative positions from location to location. This relationship is also seen in the section on the remnant deposits (page 21), invertebrate PCB concentrations can compare closely to those observed in the fish. Even though there are species differences, the overriding concern related to uptake and how it affects this discussion is the presence or absence of source(s). It is apparent from other studies, that if the source is removed, the specter of contaminated fish is also removed, regardless of species (e.g., Sloan 1999a, Parsons 2002, Skinner 1993).

Mid-point Summary

So far, we have shown in evaluating PCB concentrations in biota:

- Length/age relationships are inconsistent or sporadic, except in the younger age/size classes;
- Species differences are generally inconsistent on either wet weight or lipid adjusted bases;
- Sex differences are generally not apparent;
- Taxonomic and trophic status are not major factors;
- Seasonal influences (section on page 25) are not an overriding determinant in examining spatial patterns;
- Although not specifically addressed, factors such as reproductive stage, physiological state, and habitat requirements are also not expected to unduly influence PCB concentrations since they are related to the conditions listed above.

Basis for the ‘Species Smash’

In large ecosystems, a simple empirical approach is needed to reasonably assess the status of chemical residues at any given location. Sampling limited numbers of fish of a particular species, while concurrently attempting to address the inherent variability in the habitat, in the species (e.g., sex, age, behavior, condition, season, reproductive stage, and physiological state), and other factors related to exposure, may require such large sample sizes that a depletion of fish stocks could occur, or result in an inability to satisfy the sampling program. Recognizing that there are indeed species differences in PCB concentrations, even on a lipid basis, a simpler approach to evaluate observed conditions is desirable.

The concept of the ‘species smash’ has been developed in which all samples across several species and, in some cases, taxonomic classes are combined for any given location. Although this approach has been used in other instances on a smaller scale in an exploratory manner, the results were encouraging and showed utility for a broader context. These include both published and unpublished interpretations related to the Queensbury site (Parsons 2002), Nassau Lake and the Valatie Kill (Sloan 1999b), Love Canal drainage (Skinner 1993), St. Lawrence River (Sloan and Jock 1990), the Hudson River (Sloan 1999a, Field et al. 1996, Sloan and Field 1996, Sloan and Kane 2001), and statewide for evaluating relationships between PCBs in fish and PCBs in mink and otter (Foley et al. 1988).

In the following discussion, no ‘outlier’ has been removed and all organisms tested are used. The objective here is to simplify the spatial patterns by using all the data available to increase the robustness of the emerging relationships between locations, that produces an inescapable insight and indelible picture from the data spectrum. The presentation of the ‘species smash’ for the 1999 Hudson River sampling begins with Figure 10 and is discussed below.

Source Conditions

Examination of the overall ANOVA - Figure 10 illustrates the variation of PCBs in biota with respect to source conditions in the river in 1999. The analysis of variance (ANOVA), which was significant at $P < 0.00001$, also provides the means and confidence intervals associated with pairwise comparisons conducted as least significant difference (LSD) tests on log10 lipid based total PCB concentrations. The plotted data are provided in Table 9-A and a presentation of the multiple range tests appear in Table 9-B. The relatively wider confidence intervals for some locations in comparison to others are due to samples being comprised of small numbers and/or a single species.

The following discussion relates to the results of the ANOVA to known and suspected PCB sources proceeding from upstream to downstream as shown from left to right in Figure 10. The river mile (RM) designations are approximations and provided as interpretive aids.

RM 301 In the headwaters of the Hudson River, the PCB concentrations at Lake Sanford (RM 301) are significantly lower than those of any other location in the mainstem Hudson River sampled in 1999. This location defines the 'background' condition at this time for PCBs in the biota of the river. There are no known or documented PCBs available to biota in this part of the system.

RM 212 - 209 The next five locations represent the sampling points associated with the Niagara Mohawk Queensbury PCB site (RM 210) which is located in the pool upstream of the Sherman Island hydroelectric dam (Figure 1-B). River mile points 212 and 211.2 are the upstream reference locations. RM 210.2 is located directly across the river from the waste site about 800 feet away. Obviously this site is causing an increase in PCB concentrations in the fish in the immediate vicinity. Interestingly, the fish immediately adjacent to the site across the river largely escape the influence. At RM 209.5, a sampling point just upstream of the dam and at the Town of Queensbury water intake, contamination is discernible but much lower than at the Queensbury site itself. The four reference locations for the Queensbury site (RM 210) may be considered as 'baseline' conditions for the rest of the river. There is some PCB contamination present but it is well above the 'background' levels further upstream.

In the Queensbury reference samples and those from Lake Sanford, many of the results are reported at the analytical detection limits. In reality the concentrations may in fact be much lower. Therefore, in order to obtain a more realistic evaluation of PCB conditions, the detection limits at the laboratories need to be reduced. If data are reported with high detection conditions, the results from locations where contamination is low or non-existent are unduly inflated. This is particularly a problem where lipid adjusted data are used and at sites where the efficacy of remediation is under scrutiny.

RM 204 - 196 RM 204.2 is a historical sampling area for long term trend species from the Feeder Dam Pool upstream of the City of Glens Falls and is represented here by relatively large numbers of fish, hence the tighter confidence interval. The small numbers of smallmouth bass taken in the vicinity of the Ciba-Geigy hazardous waste site at RM 197.3 are the cause for wider confidence intervals at this location. The sample for the Ciba-Geigy site provides a reference condition for the PCB source area associated with the 'pumphouse' and the Fenimore Bridge. The General Electric Hudson Falls Plant had its major discharge point at the 'pumphouse' and consequently this location (RM 196.1) exhibits considerable contamination. Since some remediation was completed just prior to the collection of fish in 1999, it is necessary to resample this area for long term trends and to evaluate the efficacy of the remediation. The two locations directly across the river from the 'pumphouse,' RMs 196.3 and 196.2, although having lower PCB levels, may have some secondary contamination associated with the small island on the west shore. During reconstruction of the Fenimore Bridge, the hydroelectric plant on the west shore and the Bakers Falls Dam in the late 1980s and early 1990s, some contaminated materials may have been relocated. There was also a period of time that the now closed Hudson Falls Sewage Treatment Plant located just upstream of the approach to the Fenimore Bridge accepted the waste stream from the GE Hudson Falls capacitor plant. There is a possibility that the discharge to the river resulted in contributing or introducing PCB contamination to the west side of the river. Evaluation of the extent of contamination above Bakers Falls Dam will require additional sampling to better characterize, whether and where, there is a source condition off the west shore. It is interesting that even the 1974 data of Nadeau and Davis (1976) exhibited relatively high concentrations in the biota from the east side of the island associated with the west shore, which they referred to as a 'control' area. These concentrations were about an order of magnitude less than the levels observed further downstream which in relative terms is comparable to what was found in 1999.

RM 195.8 In the river reach from the point above the 004 outfall at RM 195.8 to the south end of Remnant 5 at RM 194.1, the geography and findings become more confusing and will be discussed at greater length in the section on remnant deposits (page 21).

RM 194 - 189.4 At the east channel of Rogers Island (RM 193.2) in the Village of Fort Edward, source conditions related to the sediments of the Thompson Island Pool influence contaminant conditions in the fish. PCB concentrations are at their maximum here in the Thompson Island Pool and then generally decline with distance downstream. Samples were taken from near SA13 (RM 192.1) to examine whether PCBs may be emanating from this facility. SA13 is a dredge spoil site on the bank of the river that resulted from sediment removal actions in the river from 1951 through 1979 (Malcolm Pirnie 1992). The findings indicate no clear evidence of a continuing source since the concentrations were actually less than at Rogers Island and lower than in the samples taken from the channel behind Griffin Island at RM 189.1. This latter channel has served as the principal collection location for fish to describe temporal trends in PCB contamination for the Thompson Island Pool.

RM 189.4 As indicated earlier, some locations produce comparatively lower PCB results than

others if the sampling is restricted to a single species or a small number of fish and at RM 189.4 samples were comprised of just yearling pumpkinseed from the east side of the river.

RM 189.1 - 142 From RM189.1, the channel on the west shore behind Griffin Island, and continuing downstream, concentrations decline fairly steadily to below the Federal Dam at Troy (RM 153.2) downstream of the confluence with the Mohawk River. The most significant departure from the general decline through this reach occurs at RM 185.1 where the fish concentrations rise significantly. This location features the contaminated sediment conditions associated with *hot spot* 28 which is documented as a relatively large PCB sediment reservoir (USEPA 2002). At RM 142, in the South Turning Basin at the Port of Albany, only juvenile pumpkinseed were sampled and, hence, levels are relatively low.

RM 135 - 122.1 Shad Island (RM 135) in the mainstem of the Hudson River produced concentrations in fish comparable to those just below the Federal Dam (RM 153.2). On the other hand, levels in the fish collected in the channel behind Lower Schodack Island near Schodack Landing (RM 132.7) on the east shore of the river are significantly less than those from Shad Island and from Stockport Middle Grounds (RM 122.1). The protection from the main river appears beneficial for keeping the PCB concentrations somewhat lower in the area of the hamlet of Schodack Landing. That the concentrations are as high as they are at Shad Island is some cause for concern since the drainage through the Binnen Kill originates from the rail yards in Selkirk, New York. In preparation for the 1999 sampling, the Division of Water at NYSDEC initiated a special study in 1998 focused on whether PCBs were potentially moving to the river from the Selkirk rail yards through the Binnen Kill. PCBs were detected in the drainage in the vicinity of the rail yard as measured through the use of passive sampling devices but there was no evidence that PCBs were reaching the river (Chandler Rowell, personal communication, 3/22/02, report in prep.). Perhaps, there is some need to further evaluate this potential source.

RM 113 Concentrations increase again in the vicinity of Catskill (RM 113) compared to values in the fish observed at Stockport Middle Grounds (RM 122.1). This finding underscores the potential for secondary sources in the vicinity of the mouth of Catskill Creek and follow up sampling was conducted in 2001. Samples were awaiting analysis at the time of this writing.

RM 100 - 75 Concentrations decrease again in the Tivoli Bay (RM100) and Esopus Meadows (RM88.2) areas but increase again in the vicinity of the City of Poughkeepsie (RMs 77.5, 76.8, and 75.7). This area was chosen for particular attention due to a potential source condition at the Hudson River Psychiatric Center (HRPC). The HRPC is located on a drainageway to the river that enters the Hudson River in the vicinity of Marist College on the east bank in the City of Poughkeepsie. A cleanup was undertaken in the late 1990s at the now closed hospital and the purpose of the sampling in 1999 was to gather data on whether the site was an actual source. Although there was a slight elevation in levels at (RM 76.8) and above Marist College (RM 77.5) the results were not significantly different from the general location for Poughkeepsie (RM 75.7). Unfortunately, there were no comparative samples taken before the remediation occurred. Since the site was located some distance away from the river and it was a comparatively small problem,

it is possible that it never had a discernible influence on the river (LMS 1996, 1998).

RM 73.1 The relatively low levels in fish at RM 73.1 (Blue Point) involve a small sample size of eight (8) striped bass.

RM 60 - 27 The two sampling points in the Newburgh Bay area were not different from each other (RMs 59.5 and 60) and are not much different than the Constitution Island locations (RMs 54.3 and 52.3) or Iona Island (RM 47). RMs 40.1 and 34.2 involve striped bass only and these locations were not different from the fish sampled near the Tappan Zee bridge (RM 27). The Tappan Zee Bridge fish, however, contained much lower PCB concentrations than the multiple species locations further upstream.

RM 24.8 - 13 Concentrations increase substantially at Piermont Marsh (RM 24.8). The levels, however, were less than those from the Dobbs Ferry area (RMs 23.2 and 23.1). The Village of Dobbs Ferry location (RM 23.1) did produce samples higher in concentration than the fish from Hastings-on-Hudson (RM 22.1). These results raise the issue of whether there are unknown source conditions in this area of the lower Tappan Zee.

Just downstream of Hastings-on-Hudson (RM 22.1) there is a hazardous waste site known as Harbor at Hastings. The sampling location immediately adjacent to the hazardous waste site is represented by samples collected from a small beach area near MacEachron Park (RM 21.9). Samples from an abandoned marina (RM 21.3) at the site produced organisms significantly higher in contamination than samples from the locations on either side, MacEachron Park (RM 21.9) and the North Slip (RM 21.2). Remember that the RMs as given are misleading since they do not reflect actual distances between locations which may only be separated by a few dozen yards rather than tenths or halves of miles. They are convenient numbers to designate specific locations. Refer to the accompanying figures for reference of scale (e.g., Figure 1-D). The South Slip (RM 21.1) location at the southern end of the hazardous waste site also produced samples with higher PCB levels than the samples from the North Slip. In this case, the organisms collected, vertebrate and invertebrate, describe and delineate the contamination from the Harbor at Hastings waste site.

The last location, depicted in figure 10, involves only spring collected striped bass north of the George Washington Bridge at RM 13.

Spatial Aspects Using Average Species Values - Figures 11-A and 11-B show wet weight and lipid based average values, respectively for each individual species. These figures illustrate the variability associated with average values for individual species on both wet weight and lipid basis. Note that the wet weight total PCB values for carp are substantially greater at some locations, such as Coveville and in the vicinity of the locks downstream. When expressed on a lipid basis the averages are more similar to the other species.

Plots of the ‘Species Smash’ Averages - When all the species are combined a clearer picture emerges (Figures 12-A and 12-B). On a wet weight basis (Figure 12-A), the average concentration shifts markedly from one location to another. On a lipid basis (Figure 12-B) the average concentration also shifts markedly, but the pattern is different. In both situations strong responses to source conditions are evident including through the Upper Hudson River in the vicinity of the contaminated sediments. Some areas seem to produce a greater response than others, for example the east channel of Rogers Island compared to the area around Special Area 13. The general source condition in the Upper River is a principal feature of the observed gradient. Other more localized source conditions are also observed, in particular the Harbor at Hastings site as seen by the average at the ‘Abandoned Marina’ location and the Niagara Mohawk Queensbury hazardous waste site (NiMo 1) located above Glens Falls. The more highly chlorinated ‘Aroclor 1254 +’ portion of the PCB mix is an important feature of the total PCB available at any location where contamination is elevated with the possible exception of some of the most upstream sites above the Niagara Mohawk Queensbury location. Conditions associated specifically with the yearling pumpkinseed and the remnant deposits are discussed in further detail below.

Figures 13-A and 13-B are simplified versions of the two previous figures in that the locations which had limited numbers of species, such as yearling pumpkinseed and striped bass, were removed from the depiction. The result is a generally smoothed gradient with obvious source conditions which are apparent, particularly on a lipid basis. Again, the accentuated peaks indicate the principal source conditions. In particular, note that ‘1254+’ is elevated relative to total PCB in the east channel of Rogers Island compared to the site of the original PCB discharge to the river from the ‘pumphouse’ above Bakers Falls. In addition to the local sources already mentioned, there are indications of potential added sources at Catskill, perhaps near Shad Island (although this may be due to some of the influence from above the Federal Dam at Troy), in the vicinity of Poughkeepsie, and in the area of Dobbs Ferry.

In Figure 13A-B, for the locations upstream of Bakers Falls (i.e., GE Pumphouse) only fish from the east shore were retained. Samples from the west side of the river are not included, since there were large discrepancies in some samples to the extent that the distribution of the data was more bimodal in nature and tended to confound interpretations. The data from the west side samples tended to distribute into two categories - high concentrations versus low concentrations. To simplify the graph, the west side locations were eliminated.

Spatial Trend by Regression - In the Upper Hudson River, a regression of the average PCB wet weight values excluding the remnant deposits and specialized collections (e.g., yearling pumpkinseed) exhibited a strong relationship over distance. Between the source area as shown by the ‘pumphouse’ condition to the pool above the Federal Dam (i.e., Pleasantdale area) an R^2 of 0.47 for a power fit of the data was obtained. A power fit was selected since it maximized the R^2

for the trend line shown in Figure 14-A. On a lipid basis, also using a power fit of the data, the R^2 increased to 0.82 (Figure 14-B) and smoothed the trend line by reducing the effect of the highly contaminated carp at Stillwater. The carp value generated a 'spike' in the line for the wet weight values seen in Figure 14-A. Although at much lower concentrations, the spatial gradient for '1254+' also exhibited an exponential decline away from the upstream areas. The trend lines for total PCB and 'Aroclor 1254+' tend to converge with distance downstream.

In the Lower Hudson River, similar patterns were observed as exemplified in Figures 15-A and 15-B as linear fits. In this section of the river, the starting point was below the Federal Dam in Troy and continued downstream to just above the Hastings hazardous waste site. The R^2 drops substantially on a lipid basis from 0.82 in the Upper River to 0.23 in the lower river. A similar condition was noted in the R^2 's for the '1254+' component, changing from 0.56 in the Upper River to 0.05 in the lower stretch. In relative terms the conditions in the Lower River appear to be more stable over the 150 mile course compared to the situation in the upper 50 miles. As in the Upper River the trend lines for total PCB and the 'Aroclor 1254+' component tend to converge with distance downstream, i.e., the more highly chlorinated type of PCB becomes more predominant in the total PCB mix. There is another feature in the data which emerges overall. The lipid-PCB relationship in the Lower River where there are also lower contaminant concentrations is not as strong, i.e., the R^2 on a lipid basis drops from 0.82 (Figure 14-B) in the Upper River to 0.23 (Figure 15-B) in the Lower River. On a wet weight basis, the R^2 's are similar, 0.69 and 0.75 for the upper versus lower portions of the river, respectively (Figures 14-A and 15-A).

Armstrong and Sloan (1988) noted that the lipid-PCB relationship in yearling pumpkinseed reflected a reduced correlation over the spatial gradient of PCB contamination, i.e., correlations between PCB and lipid were reduced as the concentrations in the fish decreased as a function of distance from a major source. Perhaps, some of the lower correlations noted in Table 8 and reflected above are due in part to lower exposures in less contaminated areas.

Remnant Deposits - Observations between Sites and within Sites - Since the areal extent of the sampling locations (Figure 1-D) associated with each of the Remnant Deposit sites was limited (11 sampling sites within about 1.5 miles), both smaller fish and aquatic invertebrates were targeted for collection. The results of the fish portion are included in Table 3 and the invertebrate analyses are presented in Table 4.

Coupled with the need to evaluate conditions within and between the remnant deposit sites themselves, there was the obvious influence of the 004 outfall from the Ft. Edward Plant Site and its impact on biota in the area. Although the effect of the outfall and the influence of the various deposits on the biota in the reach of the river between Bakers Falls and Ft. Edward is readily discernible in the larger graphs for the entire river (Figures 10 and 12), Figures 16-A and 16-B provide more detail for these smaller, more discrete locations. Figure 16-A illustrates the heavy influence of the 004 outfall area on the wet weight PCB concentrations in both fish and invertebrates in the vicinity of remnant 3 with concentrations declining with distance from the

outfall itself. Additionally, it shows the pattern of higher concentrations at the downstream end of each remnant deposit. The more highly chlorinated mix of PCB as represented by 'Aroclor 1254+' is also in evidence but total PCB is most heavily influenced by the lighter chlorinated compounds.

On a lipid basis (Figure 16-B), concentrations are lowest in the area immediately above 004, which is presumably representative of the PCBs emanating from the Bakers Falls area which includes the GE Hudson Falls plant site. The results are minimal compared to the high concentrations available to the biota at the 004 outfall and immediately below the outfall (i.e., the north end of Remnant 3). Overall, the concentrations in the invertebrates do not appear much different from the fish in terms of uptake. The fish at or just below the outfall are higher in concentration than the invertebrates but the two zootypes become comparable in the middle and downstream segments of the remnant 3 deposit. In the vicinity of the outfall, the fish may be exhibiting the influence of the waterborne aspect of the source condition. The invertebrates, on the other hand, may manifest more of the sediment contribution. Concentrations in both fish and invertebrates are comparable at the other locations on both the east and west banks.

The large differences in concentrations observed in the biota between the two banks underscore the influence of the 004 outfall on the east bank of the river and imply that the two sides of the river are not apparently mixing throughout this reach either in terms of biota mobility or water/sediment interactions. It is possible that the west bank was never as heavily impacted by the historical discharges, or that the pattern of contamination shifted through time, post 1973 dam removal. Whatever the reason, the 004 outfall is exhibiting the majority of the impact in this reach of the river, at least at the present time.

Also notable, in Figures 16-A and 16-B, is the tendency of the concentrations to increase between the upstream versus the downstream ends of each of the deposits. This aspect is deserving of further investigation. Is Remnant 5 having its own localized influence on the river? It is possible that the elevated levels in the south end of Remnant 5 are influenced by conditions in the northern (upstream) area above the east channel of Rogers Island. Sampling at the actual southern end of Remnant 5 was problematic due to high flow velocity and deeper water which prevented effective sampling at the downstream end of the site (Figure 1-D).

Mapping Geographic Changes in PCB

Color coded maps of concentrations throughout the river provide a visual aid to show the influence of sources on the subsequent accumulation of PCBs in biota. Figure 17-A shows the concentrations throughout the river in 1999 starting with the most upriver site at Lake Sanford in the headwaters of the Hudson River. Since many of the sampling points were close together, many of the points plot on top of one another. Figure 17-B provides better detail for those sites associated with the Niagara Mohawk Queensbury evaluation downstream to the Bakers Falls Dam. Similarly, Figure 17-C better details the Remnant sites near the Village of Ft. Edward, New York and Figure 17-D focuses on the Harbor at Hastings site near Yonkers, New York.

Liver - Standard Fillet (Edible Portion) Relationships

Select Fish Species - Several species were collected for purposes of evaluating PCB concentrations in the standard fillet (edible portion) compared to a presumed highly fatty organ (liver) which might preferentially sequester PCBs for that animal, thereby rendering the contamination less available to other organs. For this evaluation, samples of largemouth bass, smallmouth bass, striped bass, yellow perch, white perch, brown bullhead and yellow bullhead (one fish) were collected from four locations. The PCB results for the liver and the fillet analyses are summarized in Table 10. The yellow bullhead from Coveville was collected inadvertently but was included in the analyses. The PCB levels are not appreciably different from those in the brown bullhead taken from the same location.

Figure 18 depicts the average ratio of total PCBs in the livers to the PCBs in the fillets of largemouth bass and brown bullhead from the Thompson Island Pool (Griffin Island, RM 189). For both species, the average ratios are all less than one (1), that is, liver concentrations are less than the concentrations in the standard fillets for both the largemouth bass and the brown bullhead sampled from the Thompson Island Pool.

In the Stillwater Pool (Coveville, RM 176), the situation alters to some extent. The ratios for largemouth bass are greater than one (1) on a wet weight basis (Figure 19). On a lipid basis, however, the ratios again drop below one (1). For brown bullhead the liver:fillet ratios were consistently less than one (1) for both wet weight and lipid based PCB values. Figures 18 and 19 are presented separately since the relationship is simpler and easier to visualize and the condition evaluated for the Upper River is perhaps unique to some extent.

Figure 20 becomes more complex since it depicts the liver:filet ratios for all the species and the locations involved in the liver:fillet analysis. On a wet weight basis, brown bullhead ratios are less than one (1) regardless of location and as was noted for the Upper River in Figures 17 and 18. Likewise, the wet weight ratios are less than one (1) for white perch at Albany (RM 153) and Catskill (RM 112). The other species are above one and near two (2) at the Albany and Catskill locations. At Coveville, yellow perch and largemouth bass exceed three (3). In the

Thompson Island Pool, yellow perch approach eight (8), but largemouth bass are slightly less than one (1). Both the Thompson Island Pool and the Coveville locations indicate conditions inconsistent with those observed in the species mix at Albany (Federal Dam location) and Catskill.

On a lipid basis, all species produce average liver:fillet PCB ratios of less than one (1) regardless of location with the exception of the yellow perch at Griffin Island. Again, the fish in the Thompson Island Pool and Stillwater locations are perhaps reacting differently than those lower in the Hudson River.

Atlantic tomcod - This species was targeted for the 1999 sampling effort but due to its seasonal availability and life cycle it was collected in January and February of 2000. Because of its unique status and life history it is presented as a special subject and the PCB results are summarized in Table 5 for both standard fillet and liver tissues. Since it is a short-lived species, with an average age of less than a year, obtaining older individuals for analysis is difficult. Hence, only three two-year-old fish out of 22 analyzed, appear in the table. Of particular interest are the wet weight concentrations in the body compared to the liver concentrations (Figure 6-A); there are no readily apparent age differences.

On a wet weight basis there is nearly an order of magnitude difference between the two organ types with the liver concentrations being greater. On a lipid basis, however, the differences between the organs virtually disappear (Figure 6-B). In both figures note that the preponderance of the PCB mix is represented by the more highly chlorinated, "Aroclor 1254+" fraction. Of special interest, when the amounts, as total mass in micrograms (μg) of PCB, in the liver were compared to the amount in the edible portion the results were nearly identical (Table 5). The liver apparently sequesters no more PCB than the rest of the body, represented here as the standard fillet or edible portion. Unfortunately, the weights of the standard fillets for the select fish species presented above were not taken and so a similar evaluation for those samples was not possible.

Blue crab - This species of swimming crab has been of special interest for a number of years since it is highly sought recreationally and commercially, and the hepatopancreas or liver tissue has a propensity to accumulate high concentrations of PCBs relative to the rest of the body, particularly the leg muscles. Table 11 summarizes the PCB results for the 1999 collections and Figure 21-A indicate, similar to the tomcod, the high concentrations on a wet weight basis in the hepatopancreas (liver) compared to the muscle tissue. Figure 21-B, on the other hand, shows on a lipid basis the tendency for the two tissue types to equalize, although the hepatopancreas is still substantially greater in PCB concentration than the muscle. The muscle analyses themselves are more problematic due to the much lower lipid content and hence there are attendant analytical detection limit problems at these low levels and the actual differences between the two types of tissues may be confounded to some extent. Note the tendency for a greater proportion of the more highly chlorinated 1254+ to appear in the hepatopancreas compared to the leg muscles.

Of special interest are the crabs taken in association with the Harbor at Hastings (RMs 21-

22) hazardous waste site. Here, the larger, presumably more mobile and migratory adult crabs are lower in concentration than the smaller juvenile crabs which were analyzed on a whole body basis. These smaller crabs indicate to a greater degree the influence of the hazardous waste site since they are presumably utilizing and foraging over a more restricted range compared to the larger adults. Hence, the smaller crabs have much higher levels near the site at the abandoned marina (RM 21.3) compared to the crabs from the area between the marina and the MacEachron Waterfront Park (RM 21.9). The actual distance between the abandoned marina site and the location near the park is about 400 feet. The small crabs from the abandoned marina area are also higher in PCB concentration than the small crabs from the lesser contaminated portions of the site, North Slip and South Slip. PCB concentrations in the whole crabs from the other locations in the river are less than those from the Hastings hazardous waste site locations. Table 11 and Figure 20-B show that the abandoned marina crabs have about 70 percent of total PCB appearing as the more highly chlorinated mixture, 'Aroclor1254+.' This relationship is true for the crabs from most locations, except those above Newburgh (leg muscle) and at the Federal Dam. Also, it appears that the hepatopancreas in most crab samples tends to sequester more of the higher chlorinated materials compared to the leg muscles.

An experimental plan is needed to better define the tissue/exposure relationships for PCBs in this species. Blue crab also accumulate high concentrations of metals in the hepatopancreas (Sloan and Karcher 1984) which has resulted in consumption advisories. Further evaluation of heavy metals would update the database for this species and establish temporal trends for metals. Cadmium is of especial concern.

Influence of Season

In the cooperative study with NOAA, the hypothesis was that the fish should be accumulating more PCB as the season progressed due to actively feeding on contaminated food sources. Samples were analyzed on a congeneric basis by STL. The 'Aroclor' analyses on split samples were conducted by MSCL and are summarized in Table 13. As shown in Figure 22-A, the hypothesis of seasonal increase in PCB concentrations did not occur consistently on a wet weight basis. At Newburgh, concentrations in white perch did increase through the seasons but at Catskill they did not. Largemouth bass and yellow perch had lower concentrations in the fall at Catskill. In Coveville, largemouth bass increased through the fall but yellow perch PCB levels were less in the fall compared to the spring.

Generally, on a lipid basis PCB concentrations appear to decrease from spring to fall, except at Coveville (Figure 22-B). Here, they increased in levels over the seasons, particularly for yellow perch. Concurrently, the proportion of 'Aroclor 1254+' to total PCB generally declined. The exception was the white perch at Newburgh (RM60) where the composition was constant at about 56 percent '1254+'. In the Lower Hudson River, 'Aroclor 1254+' predominates the type of PCB available for accumulation and a change in the relative composition is not expected. In the Upper River, closer to source conditions, there is generally a greater amount of the lower

chlorinated materials available for trophic level transfer. A change in availability of the lower chlorinated PCBs might be postulated due to the increased biological productivity in the summer and early fall periods which may enhance the exposure of these more water soluble materials to the lower trophic levels. Higher water temperatures coupled with the increased solubility of lower chlorinated PCBs may lead to increased concentrations in the water and increased exposure to biota and resulting overall to increased concentrations in the fish relative to the higher chlorinated forms of PCBs. The relative enhancement of the lesser chlorinated PCBs is noticeable only for largemouth bass in Figure 22-B for the Coveville (RM 176) site. There may be some seasonal changes in the congeneric composition but that more complex analysis of the data remains to be done.

In addition, seasonal samples of striped bass were collected from below the Federal Dam (RM 153) (Table 14). In the spring when the striped bass enter the river on the spawning run, many of the fish congregate near the Federal Dam at Troy in the tailrace of the dam and at a hydroelectric powerhouse. Hence, the target sample of 10 fish is usually readily available. Later, as the temperatures rise in the river and flows decline, the fish move to deeper water further downstream or back to the ocean. Consequently, samples were not readily available during July and August. Some fish did reappear in September and were sampled. In October, the full complement of the targeted numbers was collected. On a wet weight basis the concentrations did increase over the sampled months but on a lipid basis they decreased. Overall, the changes in PCB composition as reflected in the percent '1254+' in Table 14 are comparable between seasons.

Striped Bass Summary

Spring collections of striped bass are a hallmark of the Long Term Sampling Project and are simply presented here for 1999 in summary (Table 15). This species and some of the other trend species will be the subject of another paper focusing on temporal changes once the 2001 results are available. Notice the lower concentrations at Esopus where major spawning activity tends to occur, but realize the sample size is greatly reduced compared to other areas. Otherwise, the downstream PCB gradient for this spring collected species is also in evidence.

Fall collections are summarized in Table 16 and feature a downstream gradient on both wet weight and lipid basis. Fall PCB concentrations are not different from those observed in the spring (Table 15) for striped bass in the section below river mile 40. The Troy fish were discussed above in the section on seasonal changes (page 25).

Special Collection from the Ciba-Geigy Site (Ponded Backwater Area)

A small special collection, the results of which are neither included in the large summary table nor plotted on the graphs, focused on the evaluation of a cleanup at another waste site (Table 12). Samples were taken from a ponded backwater area formed from an earlier stream course of the Hudson River which had become restricted behind an island and only carried moving water during exceptionally high flows. This ponded area also trapped and sequestered relatively high concentrations of heavy metals from the operations of the Ciba-Geigy plant. Subsequently, the sediments were targeted for remediation. The biotic samples were collected in 1999 prior to remediation in order to determine baseline conditions for the pond. Seven (7) smallmouth bass were collected in the river near this location and were summarized as part of the whole river gradient (page 10 of table 3). The PCB results for the fish from the backwater pond were comparable to the concentrations in the fish in the river adjacent to this location.

Other contaminants besides PCBs were evaluated, particularly heavy metals, since the waste site involved the disposal of metal paint pigments. Cadmium was relatively high in the liver tissue compared to the rest of the body. Mercury and lead were more evenly distributed. An earthworm composite sample collected along the shore of the ponded area contained high levels of cadmium, lead and mercury. Since the earthworms were not purged of gut contents there is the possibility that the results might reflect bias due to soil contamination.

Unfortunately, there was insufficient tissue to conduct organochlorine analyses on the livers and the earthworm sample, but the PCBs in the remainders of the carcasses of the fish still provided a reasonable baseline condition for comparison to the more contaminated conditions about a mile downstream. Except for some measurable DDE, the other organochlorines were close to or less than detection limits. In 2001, remediation was completed for the Ciba-Geigy site and therefore, follow up collections are planned for 2002.

In Closing

The 1999 data on the fish and other biota from the Hudson River provide an example of what happens globally, in this case riverwide, when the system is assaulted locally. Over time, the inherent patchiness in the system develops to the extent that multiple source conditions result. The separate sources are directly observable through sampling and analysis of local biota. Each source potentially deserves remedial attention.

As time passes, and 25 years have elapsed since the initiation of the Hudson River Project, the more highly chlorinated portion of the PCB mix has persisted; it remains a major component of total PCB in the fish throughout the system. Hypothetically, it will come to dominate the temporal and spatial trend of PCB concentrations in future years and the trend will shift to a more persistent, recalcitrant pattern. Concentrations will continue to decline but the rates of decline, which are already slow, will slow further.

Documented remediation coupled with adequate monitoring, shows that source removal (original discharge points and subsequent cleanup) provides net environmental benefits. Furthermore, there can be additional benefits through removing residual (contaminated sediment) sources.

After all the arguments, all the printed documents, through all this time, it appears that much is over thought and made overly complex. In the end, there is still a simple solution. It is the source(s). It appears that Mr. Occam may have a good solution with his 'razor.'

CONCLUSIONS

- PCB concentrations in fish decrease with distance away from a source.
- Exposure conditions are not homogeneous within confined stretches of the river and subsequent accumulations of PCBs in biota reflect that heterogeneity.
- Fish and other biota do reflect localized source conditions.
- In 1999, PCB concentrations in fish varied over six (6) orders of magnitude from 'background' to 'source' conditions.
- With distance downstream from the major overriding source influence in the Upper River, the relative proportion of more highly chlorinated PCB increases.
- Although concentrations shift between seasons, these changes are not consistent among different locations and species.
- If the numbers of species and the sample sizes are sufficient, it is useful to combine species, particularly on a lipid basis, into a 'species smash' to express changes in PCB concentrations as a function of source condition(s).
- There is a need to reduce the analytical detection limits, because expression of an assumed value for samples which are below the detection limits tends to inflate estimates of total PCB, particularly on a lipid basis.
- Even though the methodology may differ from one laboratory to another, interlaboratory comparisons on split samples of fish produced similar total PCB concentrations.
- PCB concentrations in the edible portion or standard fillet, particularly for lipid-adjusted values, are comparable, if not higher, than those in the liver.
- Differences in PCB concentrations between 'trophic levels' are usually neither great, nor consistent. 'Bottom feeding' as a life habit enhancing bioaccumulation is a 'red herring.'

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